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Pump-Probe Studies of Atomic Inner-Shell Photoionization and Vacancy Decay

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Collaboration:

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The U. Michigan group has developed a laser-pump/x-ray probe capability at APS beamline 7ID [1]. The Ti-sapphire-based laser system uses an oscillator (800 nm, 88 MHz, 1 nJ, 50 fs) phase-locked to the storage-ring RF and hence the laser pulses are synchronized with the x-ray pulses produced by an undulator beamline with a double-crystal monochromator. For the experiments described in this talk, an amplifier was used to produce optical pulses (800 nm, 887 Hz, 0.3 mJ, 60 fs) at low repetition rate but much higher energy. A rate of 887 Hz was chosen to match 1/3 of the x-ray pulses (2661 Hz, 87 ps) transmitted by a mechanical chopper [2] in the beamline with the storage ring running in isolated-singlet (8 mA) mode. The laser beam was focused to 30 μm diameter over a Rayleigh range of 3 mm within an effusive jet of Kr atoms. The laser intensity within the focal volume (6×10^{14} W/cm²) exceeded the intensity expected to saturate ionization of the Kr 4p electron (14 eV binding energy). Using 20-cm-long Kirkpatrick-Baez mirrors [3], the x rays were focused to 10 μm \times 16 μm with a flux 4×10^5 /pulse and sampled the central portion of the laser focal volume after a delay of 12 ns. A Si(Li) detector viewing the interaction region recorded prompt x-ray fluorescence following K-shell excitation or ionization of either neutral- or ionized-Kr. By scanning the beamline monochromator and recording the x-ray fluorescence yields, the near-edge x-ray absorption spectra were recorded concurrently for laser-on and laser-off conditions. We observed a distinct $1s \rightarrow 4p$ resonance at 14.313 keV in the laser-produced Kr^+ ions that is absent from the absorption spectrum of neutral Kr. The observed resonance agrees well with the calculated K-shell absorption spectrum of Kr^+ . This first measurement demonstrated the ability to study inner-shell processes in laser-excited atoms.

In follow-up measurements with the laser only, an electrostatic analyzer and time-of-flight methods were used to study the assembly of $\approx 10^7$ Kr ions produced by each laser pulse. The photoelectrons escape rapidly, leaving an assembly of ions that Coulomb explodes, as suggested by the Kr^+ kinetic energy distribution that extends to ≈ 80 eV. Higher charge states are also observed at energies up to ≈ 200 eV. The focused x rays provide a penetrating probe of this ionized matter with spatial resolution ≈ 10 μm and temporal resolution ≈ 100 ps. In future experiments the dynamics of the ion assembly will be studied by varying the time delay between the laser and x-ray pulses. This x-ray probe technique could be applied to studies of similar assemblies of ionized matter such as laser-ionized clusters.

Instrumentation and techniques are being developed and used that may be useful for other experiments. These include the mechanical chopper for selecting singlet x-ray pulses [2], the Kirkpatrick-Baez focusing mirrors [3], an in-vacuum laser-focusing mirror with positioning under step-motor control, and a step-motor-controlled XYZ vacuum manipulator with ≈ 1 μm resolution. The focused laser and x-ray spots are viewed with a BGO screen and CCD camera, and the two focal spots are overlapped by scanning 10- μm -thick cross hairs with the XYZ manipulator. Temporal overlap of the laser and x-ray pulses is achieved by the pulses hitting the same fast photodiode and recording with a fast oscilloscope.

The laser system is presently being upgraded with the purchase of a new amplifier and pump laser that will provide 10 \times higher pulse energy, 5 \times higher repetition rate, and variable pulse width [40 fs – 10 ps (unchirped) or 130 ps (chirped)]. The 130 ps laser mode will allow dressed-atom experiments, i.e., where the 87-ps x-ray pulses are completely overlapped with the laser pulses. This will allow studies of

the predicted ponderomotive shift of an inner-shell absorption edge. It will also allow studies of the effects of a high-field environment on the decay of inner-shell vacancy states in combination with resonance and threshold phenomena [4]. For those experiments the Kr^{q+} ion charge state yields will be measured as a function of laser intensity as the x-ray energy is scanned across threshold. Other future pump-probe experiments of interest include studies of molecular geometries, alignment, and fragmentation.

Suggestions are made for improved capabilities to perform laser+x-ray pump-probe experiments at the APS. The ability to perform such experiments depends critically on the storage-ring bunch fill pattern. Isolated-singlet mode is advantageous for the low rep-rate, high-laser-field measurements described here. The standard 24-bunch mode, with the x-ray pulses separated by 152 ns, are well-suited to many of the ultrafast x-ray diffraction experiments [1]. Experiments are also being done at beamline 7ID and other APS beamlines in which the 88 MHz laser-oscillator pulses match 1:1 with x-ray pulses when the ring is operated in 324-bunch mode. E. C. Landahl is investigating the use of extended-cavity oscillators [5] that would produce intense laser pulses at 6.5 MHz in 1:1 synchronization with x-ray pulses under 24-bunch mode. Development of improved mechanical- or x-ray-optical choppers for selecting x-ray pulses is also desirable.

High flux in the x-ray pulses is important for the low target densities of gas-phase experiments. Having 8 mA (or more!) in the isolated singlet enhances data rates. For experiments that are insensitive to energy resolution, pink-beam x-ray pulses would provide larger fluxes. Also important is the ability to microfocus the x-ray pulses while maintaining high throughput. Our initial experiments were done with KB mirrors that provide good throughput at $\approx 10\text{ }\mu\text{m}$ resolution. We would benefit from the development of focusing methods (mirrors, lenses, zone plates) giving $\approx 1\text{ }\mu\text{m}$ resolution while maintaining flux.

APS single-bunch pulses are presently $\approx 87\text{ ps}$ FWHM. The dressed-atom experiments mentioned above rely on completely overlapping the x-ray pulses with laser pulses, hence the need to operate the laser in uncompressed mode that greatly decreases the peak intensity. Decreasing the x-ray pulse width substantially, i.e., to 1–10 ps, would allow the laser pulse width to be decreased accordingly and result in higher field intensity. This is important because high-field effects are nonlinear and quite sensitive to intensity.

Finally, several of the inner-shell phenomena we wish to investigate would benefit from the use of soft x rays to perform experiments on lower-Z atoms. The inner-shell lifetimes are longer, so the resonance and threshold structure is better resolved. The cross sections are larger, and the fewer number of electrons result in simpler decay spectra. High-resolution electron spectroscopy of atomic and molecular inner shells is a powerful technique developed at third-generation soft x-ray beamlines. Consideration should be given to the development of a pump-probe capability on a new or existing soft x-ray beamline at APS like 4-ID-C.

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